

FINAL REPORT

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Unseeded Scalar Velocity Measurements for Propulsion Flows

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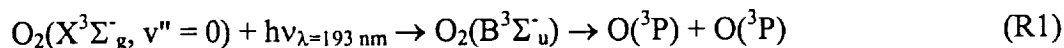
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**Concise Summary of Past and Current Research Work Sponsored by NASA-Glenn Grant
NAG3-1984:**

Unseeded Scalar Velocity Measurements for Propulsion Flows

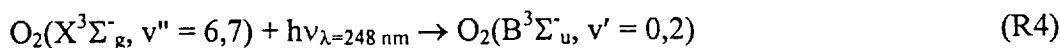
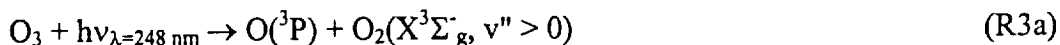
Ozone Tagging Velocimetry

Unseeded molecular tagging methods based on single-photon processes that produce long tag lines (>50 mm) have been recently developed and demonstrated by the Combustion Laser Diagnostics Group (Mechanical Engineering Department) at Vanderbilt University [1,2]. In Ozone Tagging Velocimetry (OTV) a line of ozone (O_3) is produced by a single photon from a pulsed narrowband argon fluoride (ArF) excimer laser operating at ~ 193 nm. After a known time delay, t , the position of the displaced (convected in the flow field) O_3 tag line is revealed by photodissociation of O_3 and subsequent fluorescence of O_2 , caused by a pulsed laser sheet from a krypton fluoride (KrF) excimer laser operating at ~ 248 nm. Intensified CCD camera images of the fluorescence are taken from the initial and final tag line locations thus providing unobtrusive means of establishing a velocity profile in the interrogated flow field. The O_3 lines are "written" and subsequently "read" by the following reactions:



In reaction (R1), a single photon of 193-nm light excites the ground state O_2 to the predissociative state $O_2(B^3\Sigma_u^-)$ that rapidly decomposes (at a rate of $\cong 10^{11} \text{ s}^{-1}$ [1]) into ground state (3P) oxygen atoms. The rate of formation of O_3 is governed by a three-body reaction (R2) that is slow relative to (R1). For $M = N_2$ the rate constant of the reaction $k_2 = 5.7 \times 10^{-46} \text{ m}^6\text{s}^{-1}$ under standard conditions [3]. With this rate constant, the time of formation of $\sim 63\%$ of the steady-state O_3 concentration (at STP) is $1/(k_2 n_{O_2} n_{N_2}) \cong 20 \text{ }\mu\text{s}$, where n is number density.

The displaced O_3 tag line is read by the 248-nm light sheet through reactions (R3a) and (R4):



Instantaneous OTV images of a 3×3 tag line grid are shown in Fig. 1. Fig. 1a shows an undisplaced grid (0 ms write/read delay), and Fig. 1b shows a displaced grid (1 ms write/read delay). These images were taken in an air flow from a 12.5 mm dia. nozzle with a flowrate of 20 SLPM, yielding a nozzle exit velocity of ~ 2.5 m/s where measurements were taken at ~ 25 diam. downstream of the nozzle's exit.

To predict the concentration of O_3 as a function of temperature, pressure, and time, the CHEMKIN II thermodynamic database and the SENKIN chemical kinetics solver [4] were used with a set of 119 reversible reactions, including reactions involving nitrogen oxides [5]. OTV lines are relatively insensitive to variations in air humidity or pollutant levels rendering the

technique applicable to a wide variety of experimental flow fields. In addition, studies show that an increase in pressure leads to an increase in the maximum O₃ concentration, however, elevated temperatures tend to be detrimental for the longevity of the O₃ tag [2]. Prompted by this deficiency, a new non-intrusive molecular tagging method has been developed and demonstrated at Vanderbilt University, namely Hydroxyl Tagging Velocimetry (HTV) [5,6].

Hydroxyl Tagging Velocimetry (HTV) for High Temperature Applications

The HTV technique was originally developed to be complementary to Ozone Tagging Velocimetry (OTV), that uses two narrowband, tunable excimer lasers, hence a 248 nm KrF excimer laser is used for the “read” laser in high temperature HTV work. This technique involves “writing” a line of hydroxyl radicals (OH) in a moving gas stream by photodissociating the vibrationally excited H₂O present in high temperature combustion flows. The line of OH is superimposed upon the ambient OH present. The line of “superequilibrium” OH is convected downstream and after an appropriate delay time a second laser pulse causes the OH in the moving flowfield to fluoresce, giving an image of the displaced superequilibrium OH line. The original and displaced positions of the OH line, together with the known delay time for the displaced image, can be used to determine velocity. The water vapor is dissociated by a single photon of 193 nm laser radiation to write a grid of OH lines:

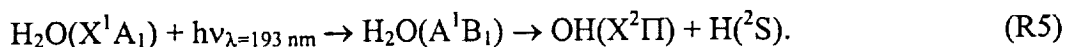


Figure 2 shows curves of potential energy surfaces for the ground (X¹A₁) and first excited (A¹B₁) electronic states of H₂O. In the read step, a light sheet from a KrF excimer laser excites the OH to fluoresce by the A²Σ⁺-X²Π (3←0) OH transition. Several rotational lines are within the tuning range of the KrF laser (from 248 to 249 nm) [7]. The strong P₁(8) line was used for this work. The resulting fluorescence from the OH tag was recorded on an ICCD camera. The original and displaced images of the OH grid were recorded to determine the relative displacement and hence the resulting velocity field.

The OH tag in Hydroxyl Tagging Velocimetry (HTV) is formed immediately by 193 nm photodissociation (R5). The formation time is only limited by the excimer laser pulse width, which is ~20 ns. This short formation time makes HTV amenable to high-speed flows where the times between “read” and “write” lasers may be less than 1 μs.

The tag line created by the ArF laser consists of both OH and H photofragments superimposed upon a reacting flowfield containing ambient amounts of OH that can obscure the detection of the tag lines. Also, the OH molecule does not exist for long periods of time in the high temperature flowfield in which it is created. To assess these issues, the SENKIN application [4] is used again to simulate a constant pressure, adiabatic system containing an equilibrium mixture of hot products from the H₂-air combustion, along with additional amounts of OH and H. An abridged version of the GRI-Mech reaction mechanism [8] is used, shortened by eliminating any reactions with carbon-bearing species [5].

The results of the SENKIN calculations are shown in Fig. 3 for H₂O photodissociation in lean and rich streams of H₂-air combustion products. Here we have assumed the initial amount of H₂O photodissociated is equal to the H₂O population in the (0,0,1) level, which is completely photodissociated by a typical laser beam (10 mJ, 0.3 mm dia.). This initial population is a

conservative estimate. Considering all the vibrational levels, we are assuming about one third of the expected photodissociation of H_2O . Note that the original OH concentrations shown in Fig. 3 are the sum of the OH resulting from direct photodissociation (R5) and the OH equilibrium concentration. For times longer than 10 ms, the OH recombines to its equilibrium concentration.

For both of the lean flames ($\phi = 0.43, 0.69$), the OH tag is produced instantaneously and lasts for nearly 1 ms. Thus in lean flames, HTV is appropriate for very high-speed to low-speed flows. In the lean flames, the OH concentration is enhanced over the amount initially produced by photodissociation. The OH concentration increases from the photodissociation occurrence to about 1 μs . Based on analysis of the production rates of OH [6], this increase in OH is due to two reactions:



In both cases, the H atoms produced in the original photodissociation step are converted to additional OH. This is confirmed by a calculation for lean H_2 -air combustion products ($\phi = 0.43$) where the initial H atom concentration was set to zero. As seen in Fig. 15, there is no increase in OH concentration when the H atoms are absent. For lean flames, the two main destruction reactions for OH are [6]:



In moderately rich flames ($\phi = 1.45$), the OH tag also persists for a long time, up to 1 ms, allowing HTV measurements over a wide range of low and high velocity flows. At $\phi = 1.45$ the main channel for OH destruction is the reaction with excess H_2 fuel:



It can be seen that HTV works quite well in both moderately lean and rich flame conditions. However, for very rich mixtures ($\phi = 4.36$), the excess fuel rapidly consumes the OH by reaction R10 and the OH tag only lasts for about 0.1 μs (Fig. 3).

Both OTV and the high temperature HTV methods are complementary in that they provide velocity measurements in low and high temperature flows simultaneously since they both use the same laser and optics system which consists of tunable ArF and KrF excimer lasers. Simultaneous ozone and hydroxyl velocimetry allows interrogation of reacting flowfields in both unreacted, cold regions (without hot, vibrationally excited H_2O) and reacted hot regions (where O_3 thermal decomposition occurs) allowing velocimetry in a wide range of experimental combustion flowfields. Figure 4 shows a typical high-temperature HTV image taken in lean H_2 -Air flame conditions.

Hydroxyl Tagging Velocimetry (HTV) for Low Temperature Applications

The KrF excimer laser accesses only relatively weak (3,0) vibrational transitions for the (A-X) OH band which are relatively weak compared to the (1,0) or (0,0) transitions that are

normally used for OH fluorescence spectroscopy. Because of the weakness of the OH lines pumped by the KrF laser, only high temperature flows have been successfully probed, because these flows have significant populations of vibrationally-excited H₂O, which have 193 nm photodissociation cross sections two or more orders of magnitude higher than ground vibrational state H₂O [3]. In low temperature HTV, the (0,0) transition of strong OH is pumped at 308 nm. This compensates for the relatively small amount of OH produced by 193 nm H₂O photodissociation at low temperatures. The ArF excimer produces a grid of hydroxyl molecules that is displaced by the flowfield and imaged by the dye laser. Figure 5 shows a schematic of the HTV system (similar to the OTV system) in current use at Vanderbilt except for the substitution of a new read dye laser. The dye laser accesses the strong Q₁(1) line at 32,474.58 cm⁻¹ (307.933 nm) in the (A-X) (0←0) OH band. Hence, as with high temperature HTV, the OH molecular grid is created via a single photon process that yields very long lines (>50 mm).

Multiline grids are used to demonstrate the feasibility of multipoint velocity measurements at room temperature conditions [9]. Figure 6 shows a 3 x 4 HTV flow-tagging grid created in a stream of room temperature air created by a 12.5 mm diam. nozzle. The measured flow rates (at 300 K, 1 atm) were ~ 205 SLPM yielding a theoretical jet exit velocity of ~28 m/s. Figure 6b shows a typical instantaneous image of the displaced HTV grid for a 30 μs delay between the firing of the ArF excimer "write" laser and the dye laser/ frequency doubler "read" laser. This is for a location immediately downstream of the nozzle's exit (1-2 diam.) near the jet's centerline. The flow proceeds from the figure's bottom right corner to the upper left corner. Figure 6a shows another image of the same HTV grid for a write/read delay time of 0 μs, thus providing initial locations for the cross points of the grid lines in the flow. Based on the initial and final locations of these cross points and knowing the time delays one can establish linear velocity vector profiles as has been previously discussed [1,2,5,6]. The initial locations of the grid lines are indicated on Fig. 6b by the superimposed initial grid (in black), and displacement vectors (in black). The cross points were numbered arbitrarily for convenient location and identification of a specific displacement vector. For example, the average length estimate for #6 displacement vector is 1.06 mm, which, when divided by the 30-μs write-read delay time, yields a measured velocity of 35.3 m/s. This experimental value is close to the theoretically expected value for the jet exit velocity of 28 m/s.

Low temperature HTV is applicable to situations where other similar methods may suffer from thermal decomposition of molecular tags (say, O₃) or insufficient vibrationally hot H₂O population fractions (high temperature HTV). Another advantage offered by low temperature HTV is the use of only one excimer laser (ArF), which does not need to be tunable because the H₂O band is a broad continuum with no distinct absorption lines. Both non-tunable excimer laser and Nd-YAG pumped dye laser lower the initial investment cost of the system as compared to two tunable narrowband excimer lasers. One could view this new low temperature HTV method as a "bridge" between situations where experimental conditions are prohibitive for either OTV or high temperature HTV. Such conditions include, but are not limited to non-reacting base flows for high-speed projectiles, or low temperature hypersonic external or internal flows.

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Publications and Presentations associated with OTV, high- and low-temperature HTV, acknowledging support from this NASA-Glenn grant:

Journal Publications

Ribarov, L. A., J. A. Wehrmeyer, F. Batliwala, R. W. Pitz, and P. A. DeBarber, "Ozone Tagging Velocimetry Using Narrowband Excimer Lasers," *AIAA Journal*, vol. 37, No. 6, pp. 708-714, June 1999.

Wehrmeyer, J. A., L. A. Ribarov, D. A. Oguss, and R. W. Pitz, "Flame Flow Tagging Velocimetry with 193-nm H₂O Photodissociation," *Applied Optics*, vol. 38, No. 33, pp. 6912-6917, November 1999.

Pitz, R. W., J. A. Wehrmeyer, L. A. Ribarov, D. A. Oguss, F. Batliwala, P. A. DeBarber, S. Deutsch, and P. E. Dimotakis, "Unseeded Molecular Flow Tagging in Cold and Hot Flows Using Ozone Tagging Velocimetry and Hydroxyl Tagging Velocimetry," *Measurement Science and Technology Journal*, vol. 11, pp. 1259-1271, September 2000.

Ribarov, L. A., J. A. Wehrmeyer, and R. W. Pitz, "Hydroxyl Tagging Velocimetry (HTV) for Low Temperature Gas Flows," *Applied Physics B* (to be submitted), December 2000.

Conference Proceedings / Papers

Ribarov, L. A., J. A. Wehrmeyer, F. Batliwala, R. W. Pitz, and P. A. DeBarber, "Ozone Tagging Velocimetry (OTV) Measurements Using Narrowband Excimer Lasers," AIAA Paper No. 98-0513, 36th AIAA Aerospace Sciences Meeting and Exhibit, Reno, NV, January 1998.

Ribarov, L. A., J. A. Wehrmeyer, R. W. Pitz, and P. A. DeBarber, "Instantaneous velocity Field Measurements of Air Flows by Ozone Tagging Velocimetry (OTV)," *OSA-Spring Topical Meetings, Laser Applications to Chemical and Environmental Analysis*, Orlando, FL, March 1998.

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Wehrmeyer, J. A., L. A. Ribarov, D. A. Oguss, and R. W. Pitz, "Water Vapor 193 nm Photodissociation for High Temperature Flow Tagging Velocimetry," Paper No. B-2-39, *Joint Meeting of the United States Sections of the Combustion Institute*, Washington, DC, March 1999.

Ribarov, L. A., J. A. Wehrmeyer, D. A. Oguss, R. W. Pitz, and P. A. DeBarber, "Development of Flow Tagging Velocimetry Methods for Low and High Temperature Experimental Flowfields," *7th Annual Graduate Student Technical Conference - ASME Region XI*, Birmingham, AL, April 1999.

L. A. Ribarov, J. A. Wehrmeyer, and R. W. Pitz, "Hydroxyl Tagging Velocimetry (HTV) for High Temperature Reacting Flowfields," Paper No. B3-57, *2000 Technical Meeting of the Central States Sections of the Combustion Institute*, Indianapolis, IN, April 2000.

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Posters

Ribarov, L. A., J. A. Wehrmeyer, F. Batliwala, R. W. Pitz, and P. A. DeBarber, "Unseeded Instantaneous Scalar Velocity Measurements of Air Flows by Ozone Flow Tagging Velocimetry (OTV)," Poster No. 118, *11th Annual Graduate Research Day*, Vanderbilt University, Nashville, TN, March 1998.

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Ribarov, L. A., J. A. Wehrmeyer, D. A. Oguss, R. W. Pitz, and P. A. DeBarber, "Development of Flow Tagging Velocimetry Methods for Low and High Temperature Experimental Flowfields," Poster No. 401, *12th Annual Graduate Research Day*, Vanderbilt University, Nashville, TN, March 1999.

Wehrmeyer, J. A., L. A. Ribarov, D. A. Oguss, F. Batliwala, R. W. Pitz, and P. A. DeBarber, "Flow Tagging Velocimetry for Low and High Temperature Flowfields," Poster No. 2-1, *10th Gordon Research Conference on Laser Diagnostics in Combustion*, Il Ciocco, Italy, June 1999.

Wehrmeyer, J. A., L. A. Ribarov, F. Batliwala, D. A. Oguss, R. W. Pitz, and P. A. DeBarber, "Unseeded Flow Tagging Velocity Methods for Low and High Temperature Flowfields," Invited

Poster presented at the "*What's Hot in Optical Sciences for 1999*" session of the *OSA/ILS-XV Annual Meeting & Exhibit*, Santa Clara, CA, September 1999.

Ribarov, L. A., J. A. Wehrmeyer, D. A. Oguss, and R. W. Pitz, "Flow Tagging Velocimetry Methods for High Temperature Reacting Flowfields," Poster No. 116, *13th Annual Graduate Research Day*, Vanderbilt University, Nashville, TN, March 2000.

Pitz, R. W., J. A. Wehrmeyer, L. A. Ribarov, D. A. Oguss, F. Batliwala, P. A. DeBarber, S. Deusch, and P. E. Dimotakis, "Unseeded Molecular Flow Tagging in Cold and Hot Flows Using Ozone and Hydroxyl Tagging Velocimetry," Poster No. 4-C07, *28th International Symposium on Combustion*, University of Edinburgh, Scotland, UK, July-August 2000.

Ribarov, L. A., J. A. Wehrmeyer, and R. W. Pitz, "Molecular Flow Tagging Velocimetry for Low and High Temperature Flowfields," Poster No. ThU58, *OSA/ILS-XVI Annual Meeting & Exhibit 2000*, Providence, RI, October 2000.

Figures

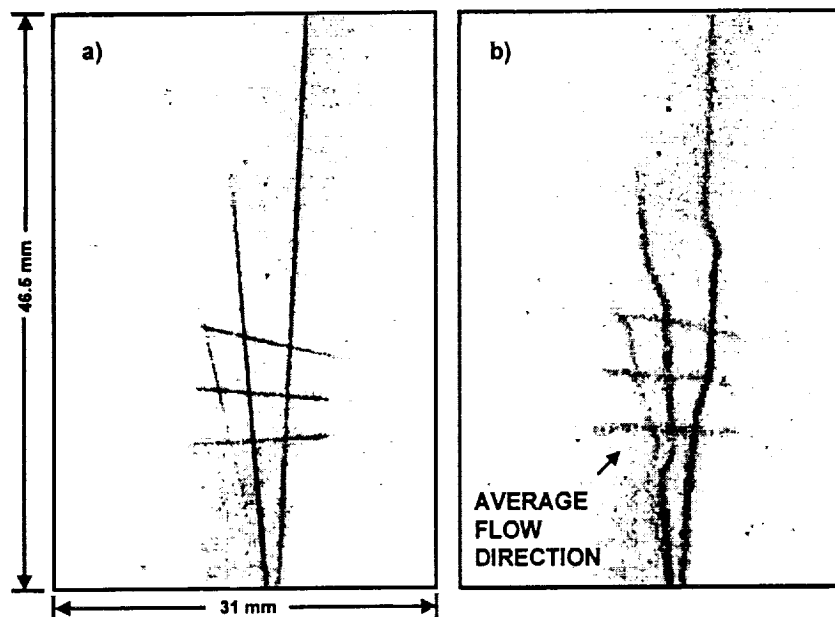


Figure 1. Instantaneous 3 x 3 OTV grid images in air flow. a) 0 ms write/read delay, b) 1 ms write/read delay [1,2].

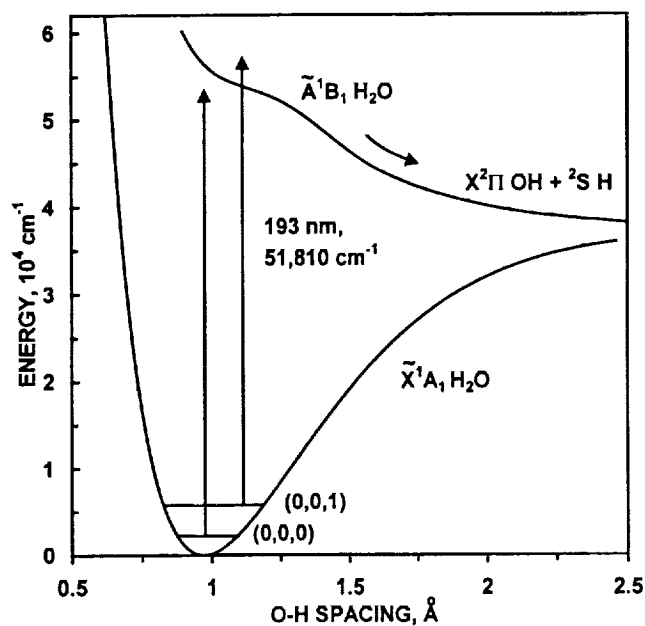


Figure 2. H_2O energy level diagram. Ground state data: Ref. 10; excited state data: Ref. 11. Both curves for equilibrium bond angle (104.5°) and equilibrium spacing for second OH bond (0.97\AA) [6].

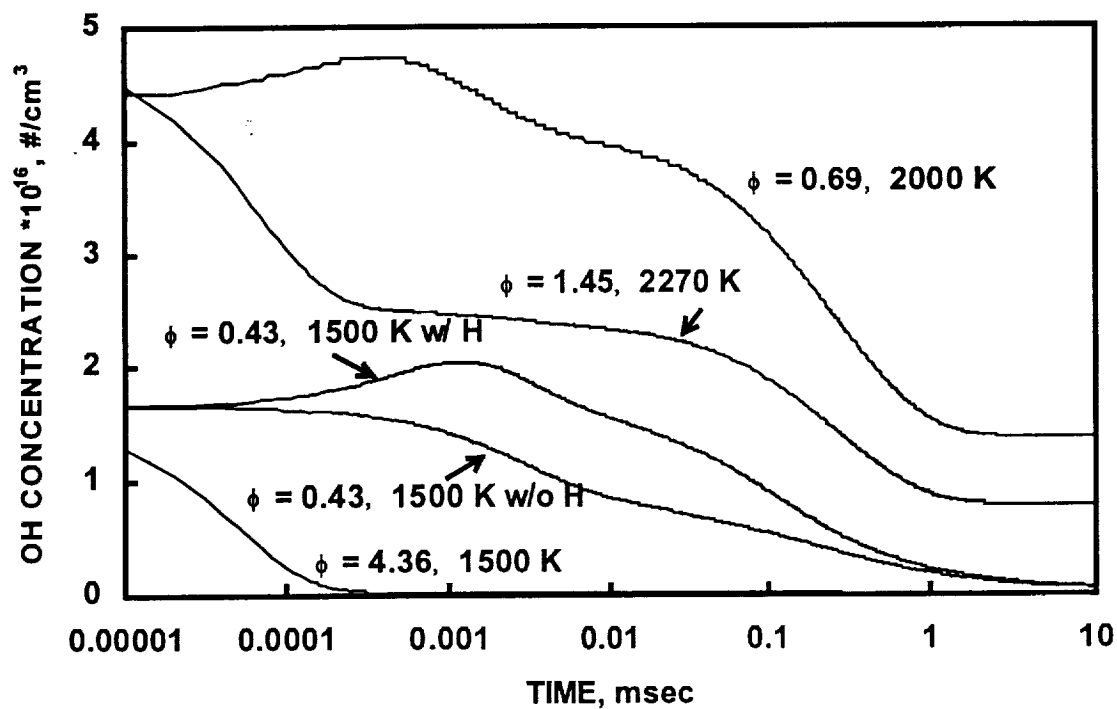


Figure 3. OH concentration vs. time for four H_2 -air flame flows, initially including OH and H photoproducts of the photodissociation of H_2O . The $\phi = 0.43$ flame modeled both with (w/ H) and without (w/o H) H photoproduct [6].

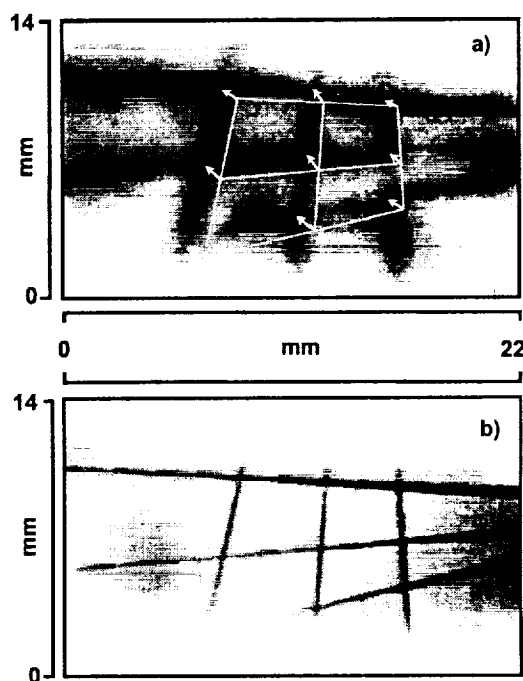


Figure 4. Single-pulse OH fluorescence images from hot H_2O photodissociation, taken in turbulent H_2 -Air diffusion flame. Write/read delays: a) $50 \mu\text{s}$, b) $0 \mu\text{s}$ [6].

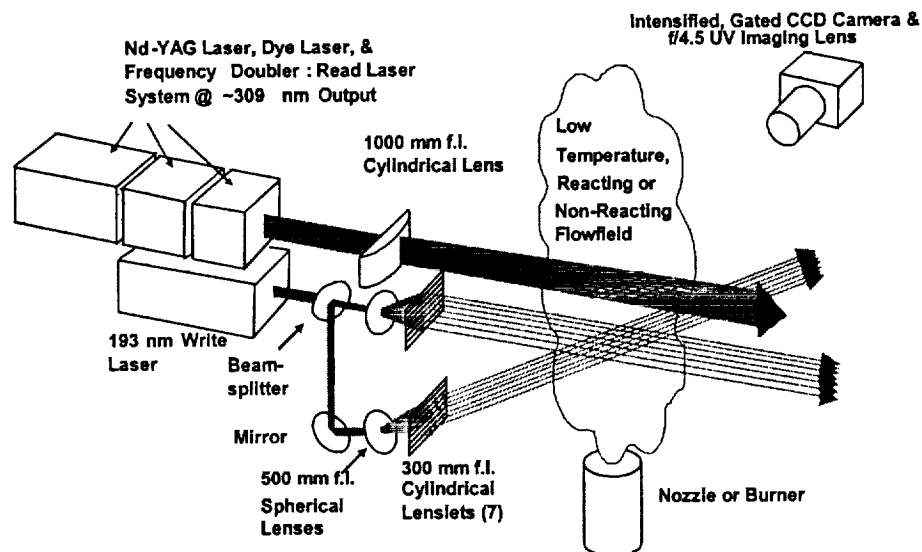


Figure 5. Schematic of low temperature HTV system, showing replacement of KrF “read” laser of previous systems [1,2,5,6] with YAG laser/dye laser/ frequency doubler “read” laser system [9].

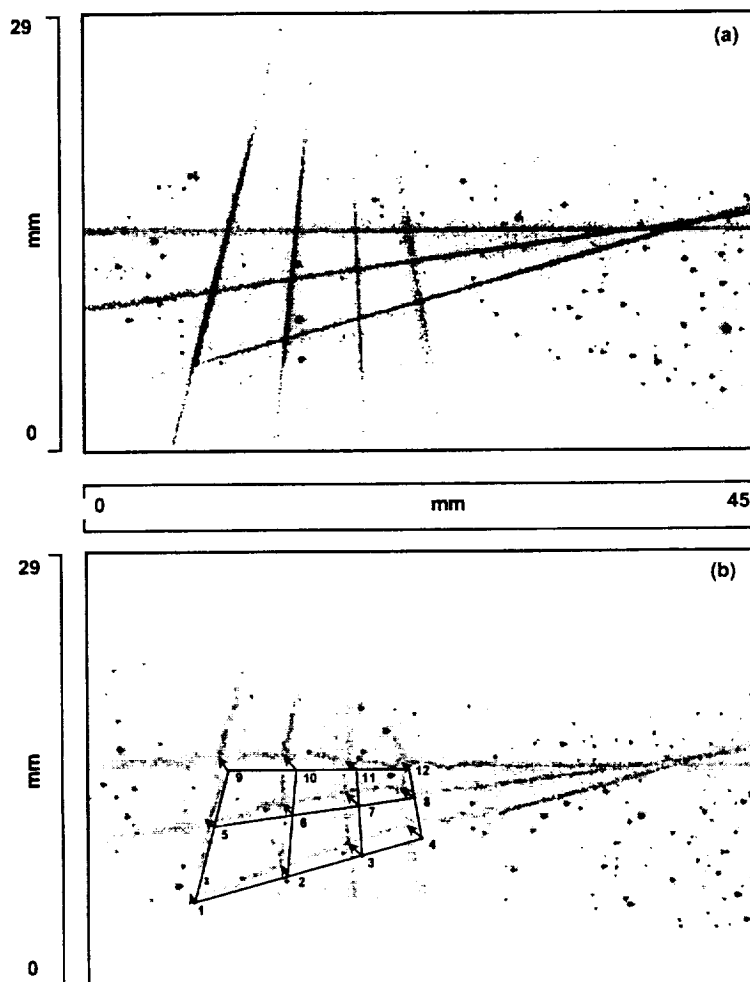


Figure 6. HTV images obtained in room temperature (297 K) airflow and 40 % relative humidity in the room air (flow moving from lower right to upper left corners in both images). Write laser: ArF excimer @ 193 nm. Read laser: frequency doubled dye @ 309 nm. Delay time between write and read lasers: (a) 0 μ s; (b) 30 μ s [9].

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